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Study on electrical switching behaviour of Ge-Se-S and Ge-$Se_{0.5}$-$S_{1.5}$ thin films

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ABSTRACT

Ternary glassy chalcogenides of Ge-Se-S and Ge-$Se_{0.5}$-$S_{1.5}$ systems were prepared by vacuum evaporation technique. Their electrical switching characteristics were examined in detail, where the resistance of the prepared thin films changed from mega to several tens of ohms. The amorphous-crystalline transition temperature (T_C) for GeSeS and $GeSe_{0.5}S_{1.5}$ thin films are around 380 K and 360 K, respectively. Due to the electrothermal mechanism a rapid transition from resistive to conductive nature was observed and the results obtained were discussed by Joule heating effect. In the prepared compositions of Ge-Se-S and $GeSe_{0.5}S_{1.5}$, sulphur atoms get their preferred bonding when connected to the two-fold coordinated selenium atoms, and the Se-S chains are cross linked by the four-fold germanium atoms. Incorporating more sulphur to Ge-Se-S system, gives the sulphur more space to adapt in the network due the smaller size of the sulphur atoms when compared to selenium. Furthermore, this method of preparation and results open-up a new approach towards the phase change memory devices that rely on easy structural transformation.

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1. Introduction

Recent advancement on amorphous systems based on Ge-Se has become significant in the field of multimedia, data storage, phase change and switching applications [1–3]. The switching effect has been fashionable in chalcogenide materials than other oxide type materials [4,5]. The chalcogenide materials show phase change behaviour that is attained by Joule heating effect [6,7]. However, a lot of research has been focused on this area to know the factual knowledge of the structural change on these materials. Quite a few sulphur based materials like $Ge_{46}S_{54}$, GeS_2, and GeS exhibited memory type electrical switching [8–10]. The inclusion of sulphur into the Se based system with fourfold coordinated Ge forms a stable glassy chalcogenide material [11]. In our previous reported data on GeSeS and $GeSe_{0.5}S_{1.5}$ compositions [12,13], thickness dependent structural disorder was studied using Raman spectra, Tauc parameter and Urbach energy. Nevertheless, in this article the research work is continued on thin films of GeSeS and $GeSe_{0.5}S_{1.5}$ to investigate the amorphous-crystalline transition temperature (T_C) followed by electrical switching of those alloys. The experimental observations concerning the dependence of electrical switching on film thickness for GeSeS and $GeSe_{0.5}S_{1.5}$ chalcogenide alloys have been discussed. This study would explore the feasibility of using these phase change materials in memory devices.

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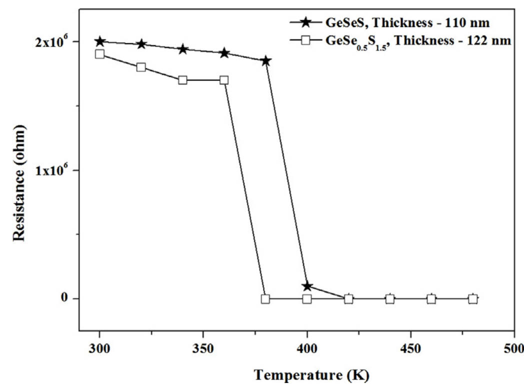

Fig. 1. Resistance as a function of temperature for GeSeS and $GeSe_{0.5}S_{1.5}$ thin films of thickness 110 nm and 122 nm.

2. Experimental details

Bulk GeSeS and $GeSe_{0.5}S_{1.5}$ glasses have been prepared by vacuum-sealed melt quenching technique. Appropriate quantities of high purity (99.99%) constituent elements of Ge, Se and S were weighed according to their stoichiometric ratio and sealed into an evacuated quartz ampoule at a vacuum of 10^{-5} Torr. Melting was performed at 1050 °C for 36 h inside a horizontal type furnace (Strohlein instruments) that allows the ampoule containing the sample to rotate with a rotational speed of 10 rpm to ensure homogeneity of the melt. Then the bulk glasses of GeSeS and $GeSe_{0.5}S_{1.5}$ were obtained by quenching the ampoules in a bath of ice water-NaOH mixture. The amorphous nature of the quenched materials is confirmed by X-ray diffraction (XRD) analysis using PANalytical X'Pert PRO (Cu as target and Ni as filter, λ = 1.5406 Å).

Thin films of GeSeS and $GeSe_{0.5}S_{1.5}$ were deposited by thermal evaporation technique (HINDHIVAC 12A4D) on cleaned glass substrates held at room temperature in a vacuum of 2×10^{-5} Torr. Three different thicknesses 110, 200, and 710 nm of GeSeS thin films were deposited, and for $GeSe_{0.5}S_{1.5}$ films the thicknesses were 122, 297, 420 and 590 nm. The substrates were located at a distance of 10 cm vertically above the source. During each deposition, the bulk material was completely evaporated at a fast rate so that no residual alloy would remain. This procedure is necessary to ensure homogeneity due to preferential evaporation of the volatile sulphur and selenium. The thickness of the films was measured using the surface roughness tester (SJ-210, MITUTOYO). For the lower thickness film of 110 nm (GeSeS) and 122 nm ($Ge\text{-}Se_{0.5}\text{-}S_{1.5}$), the resistance measurement were carried out using Keithley 590 at different temperatures using Lakeshore 335 temperature controller in the range of 300–400 K with heating rate of 5 K/min. During the measurement, the samples were heated under vacuum of 10^{-3} Torr using Janis ST-100H cryostat. X-ray diffraction analysis was made for 110 nm (GeSeS) and 122 nm ($Ge\text{-}Se_{0.5}\text{-}S_{1.5}$) thin films at 300 K and 400 K to identify the amorphous-crystal phase change. Other several characterizations such as differential scanning calorimetry, X-ray diffraction, energy dispersive X-ray analysis attached to scanning electron microscopy, photo-acoustics, transmittance and Raman spectra are reported in earlier publications [12,13].

The electrodes were prepared in sandwich geometry using aluminium as top and bottom electrodes for a thickness of 100 nm. The electrodes were evaporated through a suitable mask and they were used to perform the I–V characteristics to study the electrical switching behaviour of GeSeS and $GeSe_{0.5}S_{1.5}$ thin films. The I–V curves were recorded using a Keithley source-meter (Model 2410[c]) controlled by LabVIEW 6i (National Instruments). For the I–V measurements using the sandwiched electrodes, a constant current of few mA was applied and the corresponding voltages developed were observed. All the measurements were repeated at different portions of a thin film in order to check the reproducibility, and the error was about ±2%. The standard deviation method has been used for error analysis of the switching voltage measurements.

3. Results

The resistance as a function of temperature is shown in Fig. 1 for the film of thickness 110 nm (GeSeS) and 122 nm ($GeSe_{0.5}S_{1.5}$). At room temperature of 300 K the figure displays a high resistance of the film characterizing the amorphous nature. With the raise in temperature from 300 K, the resistance is found to decrease exponentially and an abrupt drop in resistance was noted around 360 K and 380 K for $GeSe_{0.5}S_{1.5}$ and GeSeS thin films respectively. This temperature at 360 K and 380 K is known as the amorphous-crystalline transition temperature (T_C). The high electrical contrast of four orders of magnitude between the amorphous and crystalline states of the film and the fast transition in the resistivity could be utilized for high-speed memory devices. Fig. 2 shows the X-ray diffraction pattern for (a) GeSeS thin film of thickness 110 nm at 300 K of temperature, (b) GeSeS thin film of thickness 110 nm at 400 K of temperature, (c) $GeSe_{0.5}S_{1.5}$ thin film of thickness 122 nm at 300 K of temperature and (d) $GeSe_{0.5}S_{1.5}$ thin film of thickness 122 nm at 400 K of temperature. At 300 K in both the compositions no any Bragg's peak were found indicating the amorphous nature (Fig. 1a & c). From Fig. 1 it is known that a phase change was observed at 360 K and 380 K for GeSeS and $GeSe_{0.5}S_{1.5}$ composition. These results are in well agreement with results obtained from the XRD which indicates the presence of Bragg's diffraction peak at 400 K (Fig. 2b & d). Fig. 2b

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